Synthetic magnetic rare-earth Dy-Y superlattices

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The long-range helimagnetic order in synthetic rare-earth Dy-Y superlattices has been studied. The magnetic spiral of Dy maintains its coherence in a $(Dy_{38\,Å}\text{-}Y_{38.6\,Å})_{80}$ superlattice, but not in a $(Dy_{38\,Å}\text{-}Y_{120.4\,Å})_{80}$ superlattice where the intervening Y layers are 42 atomic planes thick. Furthermore, the spiral periodicity shows a markedly different temperature dependence from that of bulk Dy, including a smaller range of variation and a "lock-in" at low temperature.

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The recent development of advanced metal molecular beam epitaxy (MBE) techniques has led to the synthesis of single-crystal magnetic rare-earth Gd-Y superlattices with coherent interfaces and chemically sharp boundaries.[1] These superlattices are ordered in long-range ferromagnetic and antiferromagnetic (antiphase domain) spin structures depending on the intervening Y layer thickness.[2,3] The long-range magnetic correlations have been interpreted on the basis of coherent propagation of the Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction through the nonmagnetic Y medium.[2–4]

Studies have now been extended to Dy-Y superlattices. Bulk Dy orders below a T_N of 178 K in an antiferromagnetic basal plane spiral structure. The exchange energy (which favors helimagnetic order) competes with the magnetoelastic energy (which favors ferromagnetic order) and the magnetic structure is eventually driven ferromagnetic through a first-order transition at a T_c of 90 K.[5] Two aspects are of primary interest here. The first concerns whether the RKKY interaction across the Y layers can also produce coherence of the Dy spiral in the superlattice. Evidence for such a long-range, coherent spiral structure was reported by Salamon *et al.*[6] The second regards the modification of the Dy spiral in a superlattice. In bulk Dy, the spiral structure is known to be strongly affected by the elastic strain, as shown by earlier pressure-dependent studies.[7] The fact that the coherency strain accommodated by the Dy layer can be fine tuned by adjusting the superlattice parameters further raises the interesting possibility of tailoring the spiral periodicity via the interfacial strains.

This work reports the results obtained on both single-layer Dy films and Dy-Y multilayers. Two superlattices were examined here with Y layer thicknesses of 14 and 42 atomic planes in each bilayer period and Dy layer thicknesses of 13 atomic planes each. Both superlattices exhibited comparable structural coherence according to x-ray diffraction analysis. However, neutron diffraction measurements have shown that the magnetic spiral coherence is greatly diminished in the sample with the thicker intervening Y layer. These results suggest that the magnetic correlation decreases with increasing Y layer thickness, and that the effective interaction range is less than 40 atomic layers. Furthermore, the temperature dependence of the Dy spiral periodicity is markedly different from that of the bulk and tunable by the superlattice wavelength.

The Dy single-layer and multilayer crystals were prepared by metal molecular beam epitaxy techniques.[1,8] Highly uniform films over an area of 2 in. diameter have been routinely made and are particularly suitable for the neutron scattering studies. Dy was evaporated from an effusion cell held at ~1300 °C whereas Y was deposited from an electron beam source. *In situ* RHEED studies indicate a highly smooth film surface with a quality at least comparable to that of Gd-Y superlattices.

The structure and coherence of the crystals were measured by x-ray diffraction. A triple-axis diffractometer was used with a Cu rotating anode x-ray source. With a Ge(111) monochromator, the instrumental resolution is 0.0008 $Å^{-1}$ full width at half-maximum (FWHM). Longitudinal scans about the (0002) Bragg reflection are plotted in Figs. 1(a) and 1(b) for the $(Dy_{38\,Å}\text{-}Y_{38.6\,Å})_{80}$ and $(Dy_{38\,Å}\text{-}Y_{120.4\,Å})_{80}$ superlattices, respectively. Judging from the narrow widths of the Bragg peak and satellite harmonics, both superlattices have similar coherence lengths over 1000 Å. The FWHM of the rocking curves perpendicular to the plane is 0.2° The asymmetry of the amplitudes of positive and negative harmonics are caused by the interfacial strain. The fact that most negative harmonics are nearly unobservable in Fig. 1(b) is evidence for coherency strain of a larger amplitude accommodated by the Dy lattices when separated by a thicker Y layer.

Magnetic measurements by the Faraday method showed that the first-order transition temperature systematically decreases with decreasing Dy layer thickness. Specifically, the first-order transition of a 5000-Å-thick Dy film epitaxially grown on Y is essentially identical to bulk Dy. In reducing the thickness to 200 Å, T_c decreases to 25 K. For a 76-Å-thick Dy film, the first-order transition no longer exists. Similarly, for both superlattices, no evidence for a first-order transition was found. The results are certainly related to a strongly modified magnetostrictive behavior for thin Dy films constrained by the adjacent Y layers. The in-plane lattice mismatch between Dy and Y is 1.6%, substantially higher than that for Gd-Y. We believe that the expansion of the Dy in-plane lattice in matching with Y reduces the tendency of developing magnetoelastic strain in the ferromag-

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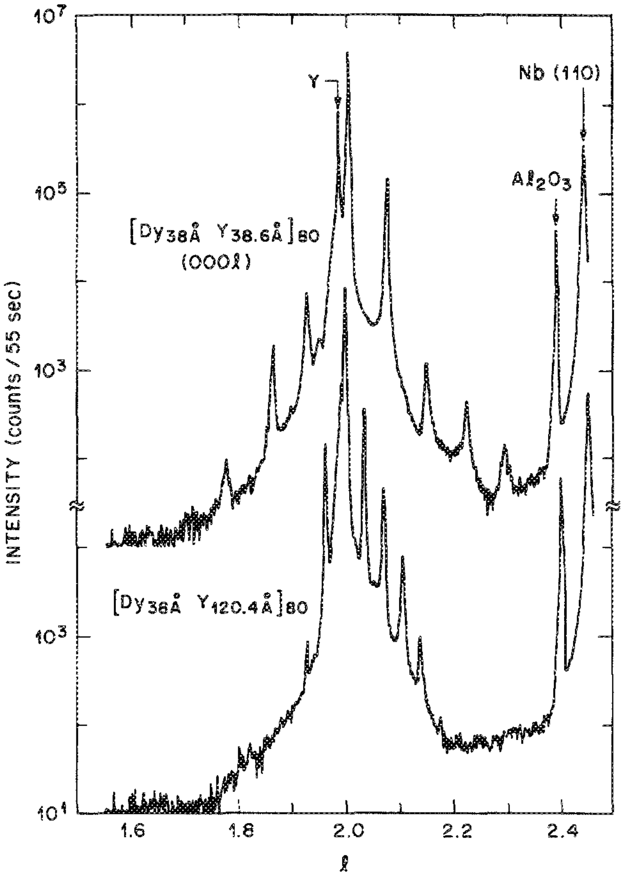

FIG. 1. High-resolution x-ray scans about the (002) reflection for superlattices of (a) $(Dy_{38\,Å}\text{-}Y_{38.6\,Å})_{80}$ and (b) $(Dy_{38\,Å}\text{-}Y_{120.4\,Å})_{80}$.

netic state. Consequently, the spiral structure remains relatively stable at low temperature.

The magnetic spiral coherence of these two Dy-Y superlattices was studied by neutron diffraction at the High Flux Beam Reactor of Brookhaven National Laboratory. The spectrometer was aligned in the parallel configuration with the following sequence of collimations and monochromators: 60′-pyrolytic graphite (PG)(002)-40′-PG(002)-15′-sample-10′-PG(002)-10′-detector. Longitudinal scans were taken with the scattering vector **Q** normal to the film. With an incident neuron wave vector $k = 1.55$ Å^{-1}, the instrumental Q resolution was measured to be ~ 0.007 Å^{-1} FWHM near the (0002) reflection.

For the $(Dy_{38\,Å}\text{-}Y_{38.6\,Å})$ sample, a simple antiferromagnetic spiral with the Dy moments lying in the basal planes was observed with a T_N slightly lower than that of the bulk. The coherence of the Dy spiral was measured to be greater than 600 Å, extending about eight bilayer periods. The results are generally in agreement with the work of Ref. 6. The temperature dependence of the positions of the primary magnetic satellites about the (0002) reflection is plotted in the insert of Fig. 2. In order to calculate the wave vector associated with the magnetic spiral in the Dy layers, a turn angle of 50° per plane in the Y layers was assumed.[9,10] The resultant Dy spiral wave vector that is consistent with the observed primary magnetic satellite positions is plotted as a dashed curve in Fig. 2. (The effects of compositional and strain modulations were neglected in the calculation.) Note that at about 45 K the spiral wave vector locks to a value of 0.224 Å^{-1}. This is in marked contrast to the behavior of the spiral wave vector in bulk Dy, also plotted in Fig. 2. The high-resolution bulk Dy data were recently obtained for a single crystal[11] and are in agreement with the earlier work of Wilkinson *et al.*[12] In bulk Dy the spiral wave vector changes continuously with temperature between T_N and T_c, unlike the case of bulk Ho or Er, where discrete wave-vector lock-ins are observed.[13]

A representative diffraction profile at 100 K is shown in Fig. 3(d). Well-resolved harmonics with a wave vector corresponding to the bilayer period ($2\pi/0.083$ Å^{-1}) appear about the (0002) reflection as well as the primary magnetic satellite whose position is indicated by the vertical arrow.

The second superlattice sample $(Dy_{38\,Å}\text{-}Y_{120.4\,Å})$, on the other hand, behaved quite differently. The coherence of the magnetic satellites is over a distance of only about 185 Å (or 1.2 bilayers) at 70 K and 260 Å (or 1.7 bilayers) at 10 K. The coherence lengths were determined by deconvoluting the observed primary magnetic satellite width and the in-

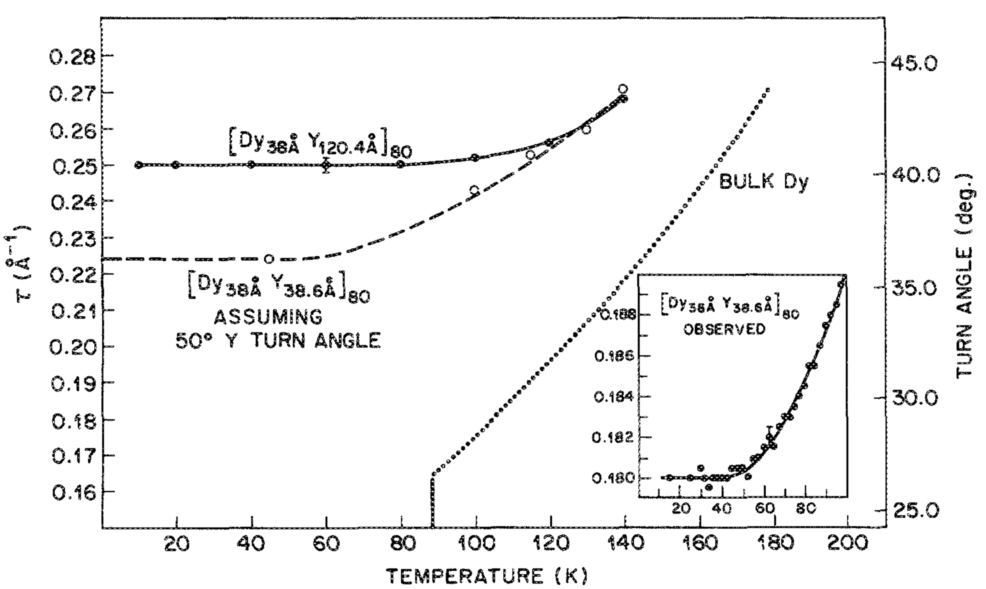

FIG. 2. Temperature dependence of the spiral wave vector of Dy in bulk Dy crystal, $(Dy_{38\,Å}\text{-}Y_{38.6\,Å})_{80}$, and $(Dy_{38\,Å}\text{-}Y_{120.4\,Å})_{80}$ as determined by neutron diffraction. The inset shows the observed wave vector of the magnetic satellites $(Dy_{38\,Å}\text{-}Y_{38.6\,Å})_{80}$ about (0002) as described in detail in the text.

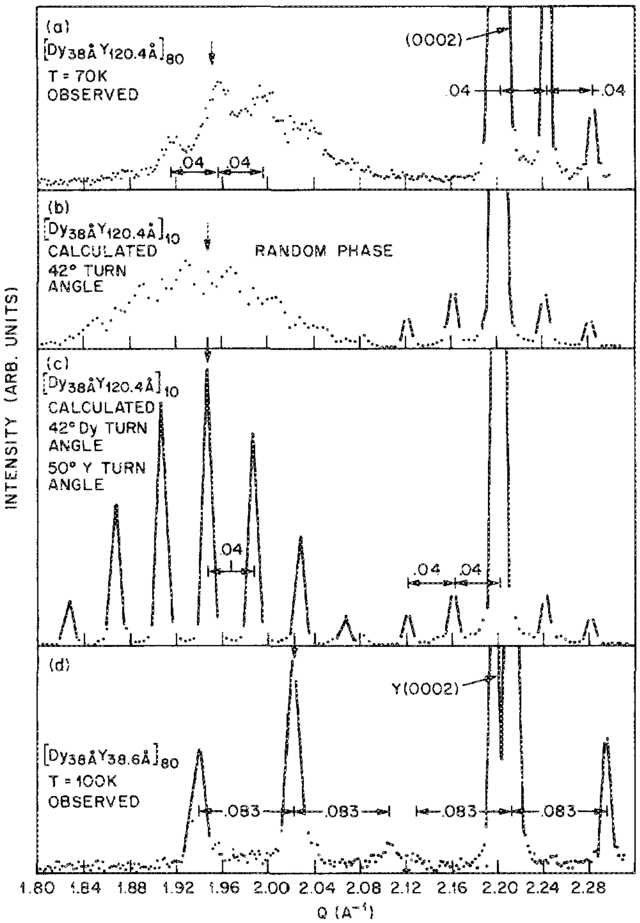

FIG. 3. Diffraction profiles about (0002) for $(Dy_{38\,Å}\text{-}Y_{120.4\,Å})$: (a) the observed at $T = 70$ K; (b) the calculated for a random phase angle across the Y; (c) the calculated for a 42° Dy turn angle and 50° Y turn angle; (d) is the diffraction profile for $(Dy_{38\,Å}\text{-}Y_{38.6\,Å})_{80}$ observed at $T = 100$ K.

strumental width. Note the broad satellite structure for this sample shown in Fig. 3(a). The vertical arrow indicates the primary magnetic satellite position as determined from that of the primary magnetic satellite about (0000), where the diffracted magnetic intensity is enhanced and therefore more accurately measured.

Figure 3(b) is a plot of the diffraction profile calculated for a $(Dy_{38\,Å}\text{-}Y_{120.4\,Å})_{80}$ sample assuming a 42°/plane Dy turn angle but a random angular phase between spirals in adjacent Dy layers. Figure 3(c) is the diffraction profile that would be expected for a $(Dy_{38\,Å}\text{-}Y_{120.4\,Å})$ superlattice which is coherent across ten bilayer periods assuming a 42°/plane Dy turn angle and a 50°/plane turn angle in the Y. The vertical arrow again indicates the position of the primary magnetic satellite. The similarity of the data in Fig. 3(a) with the calculated profile in Fig. 3(b) indicates a greatly diminished spiral coherence across the Y layers.

In Fig. 2 is also plotted the observed wave vector of the primary satellite for the $(Dy_{38\,Å}\text{-}Y_{120.4\,Å})$ superlattice versus temperature [measured about (0000)]. The wave vector varies over a smaller range and locks at about 90 K. The calculated Dy spiral wave vector for this superlattice given a 50 °Y turn angle is almost identical to that of the observed primary magnetic satellite at 70 K as can be seen by comparing Figs. 3(a), 3(b), and 3(c).

In summary, the results suggest that the coherence length of the Dy spiral across the intervening Y layers decreases with increasing Y layer thickness and is reduced to less than two bilayers for a Y spacing of 42 atomic planes. A calculation based on the RKKY coupling mechanism has been recently made[4] for two parallel planes of Dy that are separated by a nonmagnetic Y medium. Briefly, the calculation shows that a coherent spiral order will be formed across the Y layers, simply because the exchange field of a Dy layer at least 2 atomic planes thick has both x and y components in the plane. The successive Dy spirals maintain the chirality with a pitch angle repeating about every 7.2 atomic layers of Y, corresponding to $2\times 2\pi/q_{\max}$, where $q_{\max}$ is the wave vector at the maximum of the generalized susceptibility of Y, which is equal to $0.28(2\pi/c)$.[9,10] It appears that this type of mechanism could be sufficient to account for the coherent spiral order in the Dy-Y superlattice system, as it does for ferro- and antiferromagnetic order in the Gd-Y superlattice system.

The authors would like to acknowledge valuable discussions with F. J. DiSalvo, D. B. McWhan, Y. Yafet, and J. J. Rhyne. Work at Brookhaven National Laboratory was supported by the division of Materials Sciences, U.S. Department, under contract DE-AC02-76 CH00016.

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